



PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Takashi Hiraga et al.

Group Art Unit: 1774

Application No.: 09/913,315

Examiner: J. Gray

Filed: August 10, 2001

Docket No.: 110345

For: MODIFICATION METHOD OF SURFACE LAYER OF MOLDED RESIN ARTICLE
AND MODIFICATION APPARATUS OF SURFACE LAYER OF MOLDED RESIN ARTICLE

DECLARATION UNDER 37 CFR § 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Toshiko MIZOKURO, a citizen of Japan, hereby declare and state:

1. I am currently employed by National Institute of Advanced Industrial Science and Technology (AIST), an Assignee of rights in the above-identified U.S. patent application, and have been employed by National Institute of Advanced Industrial Science and Technology for four years.

2. I reside at National Institute of Advanced Industrial Science and Technology (AIST),
1-8-31 Midorigaoka, Ikeda, Osaka 563-8577 Japan.

3. I received a Ph. D. in Chemistry from Graduate School of Engineering Science, Osaka University in March 23, 2001.

4. I have been employed by National Institute of Advanced Industrial Science and Technology (AIST) since April 1, 2001.

5. I have been engaged in Photonics Research Institute in Fabrication of polymer opto-electronic devices since April 1, 2001.

6. I have reviewed and am familiar with the disclosures of the above-identified U.S. patent application and U.S. Patent No. 3,783,011 to Chauffoureaux.

7. I conducted comparative experiments that demonstrate the advantages of saturated sublimation pressure conditions. The results of the comparative experiments are accurately presented in Experiments 1 through 3 below.

Experimental 1

Two plates made of polycarbonate (PC), measuring approximately $10 \times 20 \times 0.5 \text{ mm}^3$ and $10 \times 20 \times 1 \text{ mm}^3$, were loaded in a $\phi 12 \text{ mm}$ (inside diameter) glass tube with 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)4H-pyran (DCM; Aldrich Chemical Co., Inc.; 0.005 g) and a foam polyurethane board (approximately $5 \times 5 \times 3 \text{ mm}^3$). These were used without further purification. The pumped pressure in the tube was decreased using a turbo molecular pump (Varian Vacuum Products). After reaching a pressure of $\sim 2 \times 10^{-6} \text{ Pa}$, the glass tube was sealed by melting to form an ampoule. The ampoule was 20 cm in length and was heated

at 135 °C for 72 h. (During heat treatment, residual gas got out of the form polyurethane, therefore, pressure of DCM vapor did not reach saturation vapor pressure.) After the heat treatment, the ampoule was cooled slowly over 2 h. We picked up the PC plates and confirmed that DCM was not dispersed uniformly in the PC plates. DCM was hardly dispersed one PC plate located farer from DCM. Furthermore, DCM was not dispersed uniformly into the other PC plate but dispersed gradually from one edge to the other; DCM was dispersed into one edge of the plate, but DCM was hardly dispersed into the other edge.

On the other hand, similar experiment with one alteration (without foam polyurethane board) was performed. After the heat treatment, we found that DCM was dispersed uniformly (without concentration gradient) into the two PC plates.

Experimental 2

A poly(*p*-phenylene vinylene) (PPV) film with the thickness of 500 nm, formed on a glass substrate (measuring approximately 10×20mm²), was loaded in a φ12 mm (inside diameter) glass tube with 1,4-bis(ethylamino)-9,10-anthraquinone (Solvent Blue 59; Aldrich Chemical Co., Inc.; 0.005 g). These were used without further purification. The pumped pressure in the tube was decreased using a turbo molecular pump (Varian Vacuum Products). After reaching a pressure of ~2×10⁻⁶ Pa, the glass tube was sealed by melting to form an ampoule. The ampoule was 20 cm in length and was set at 150 °C for 0.5 h. At that time, pressure of Solvent Blue 59 vapor did not reach saturation vapor pressure. We found that Solvent Blue 59 was not dispersed uniformly in the

PPV film but dispersed gradually from one edge to the other; Solvent Blue 59 was dispersed into one edge of the film, but Solvent Blue 59 was hardly dispersed into the other edge.

On the other hand, after the further heating of the ampoule at 150 °C for 24 h, we found that Solvent Blue 59 was dispersed uniformly into the PPV plates.

Experimental 3

A test sample made of poly(methyl methacrylate) (PMMA, Mitsubishi Rayon; Mw, 5~6 million), measuring approximately $3 \times 4 \times 40 \text{ mm}^3$, was loaded in a $\phi 12 \text{ mm}$ (inside diameter) glass tube with ethyl heptafluorobutanoate (EH, 0.02 g). EH (Tokyo Kasei Kogyo Co., Ltd., Tokyo, Japan) were selected as model in the present method and showed lower refractive index than PMMA used as the matrix rod. These were used without further purification. The pumped pressure in the tube was decreased using a turbo molecular pump (Varian Vacuum Products). During pumping, these organofluorine compounds were cooled with liquid N_2 to decrease the vapor pressures at room temperature under reduced pressure. After reaching a final pressure of around 10^{-6} Pa , the glass tube was sealed by melting to form an ampoule. The ampoule was 50 cm in length and was set at 135 °C for 24 h. During heat treatment, EH was entirely evaporated in the ampoule. After treatment, the ampoule was cooled slowly over 2 h. We picked up the sample and confirmed that EH did not dispersed in the PMMA rod.

On the other hand, similar experiment with two alterations (EH, 2 g; the length of the ampoule was 10 cm) was performed. In heat treatment, we confirmed that liquid EH existed in the

ampoule. The resulting sample was observed that EH could dispersed in the PMMA rod with a layer structure. We believe that the latter condition reached saturation vapor pressure, but the former condition did not reach saturation vapor pressure.

8. I hereby declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine and/or imprisonment under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or patent issuing therefrom.

Date: July 19, 2005

Toshiko Mizukuro

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